

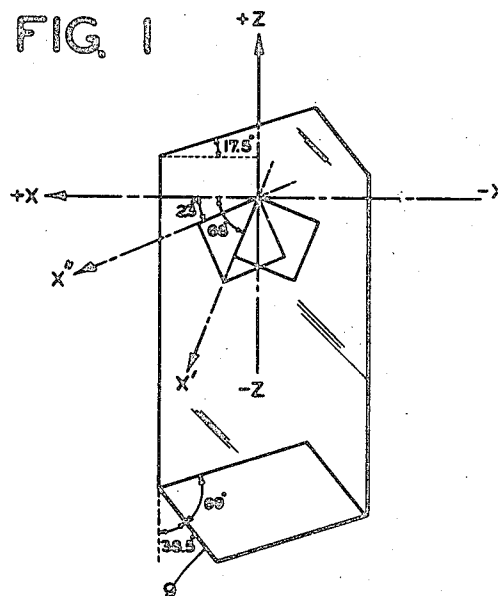
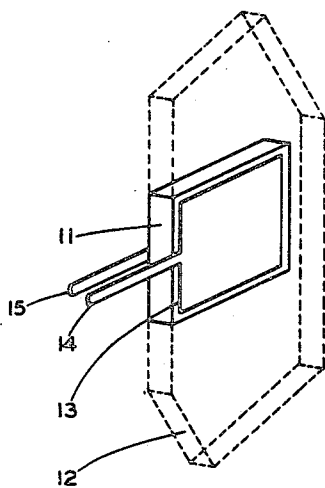
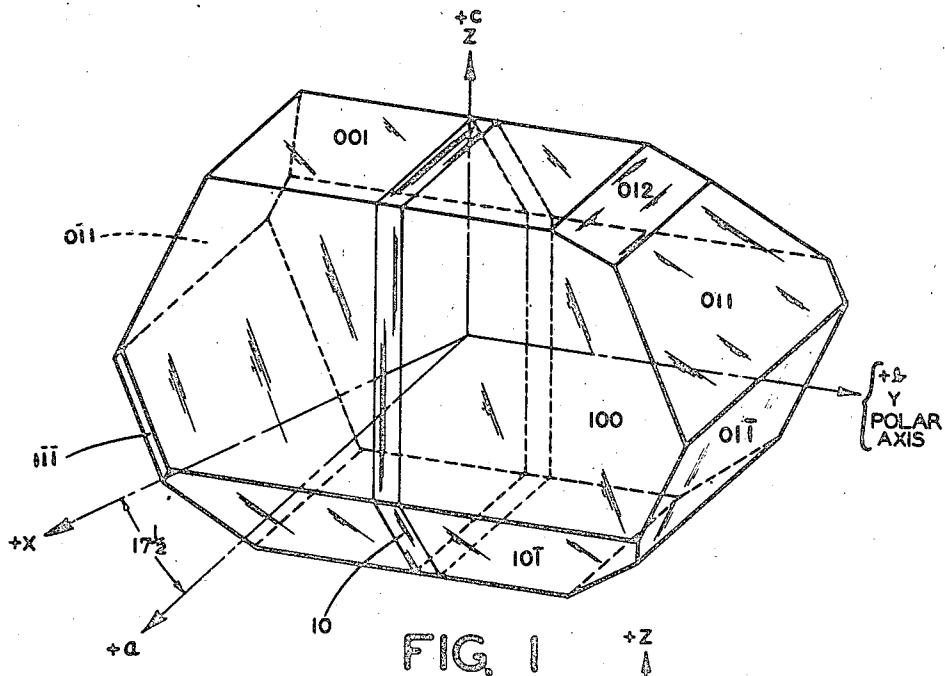
Dec. 6, 1949

H. JAFFE
PIEZOELECTRIC CRYSTAL

2,490,216

Filed June 17, 1947

2 Sheets-Sheet 1



INVENTOR.
HANS JAFFE

BY

Elias J. Hyde
ATTORNEY

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2 Sheets-Sheet 2

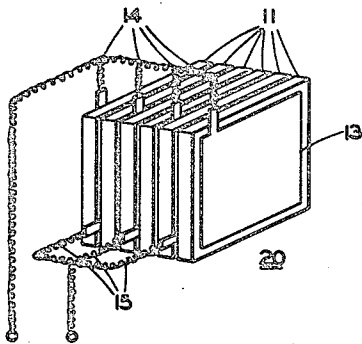


FIG. 4

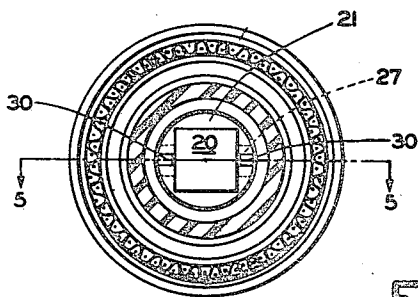


FIG. 6

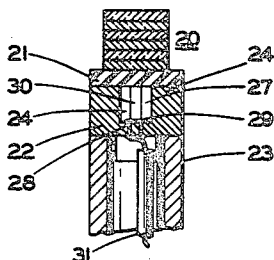


FIG. 7

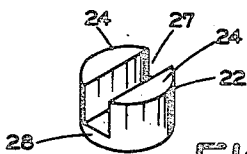


FIG. 8

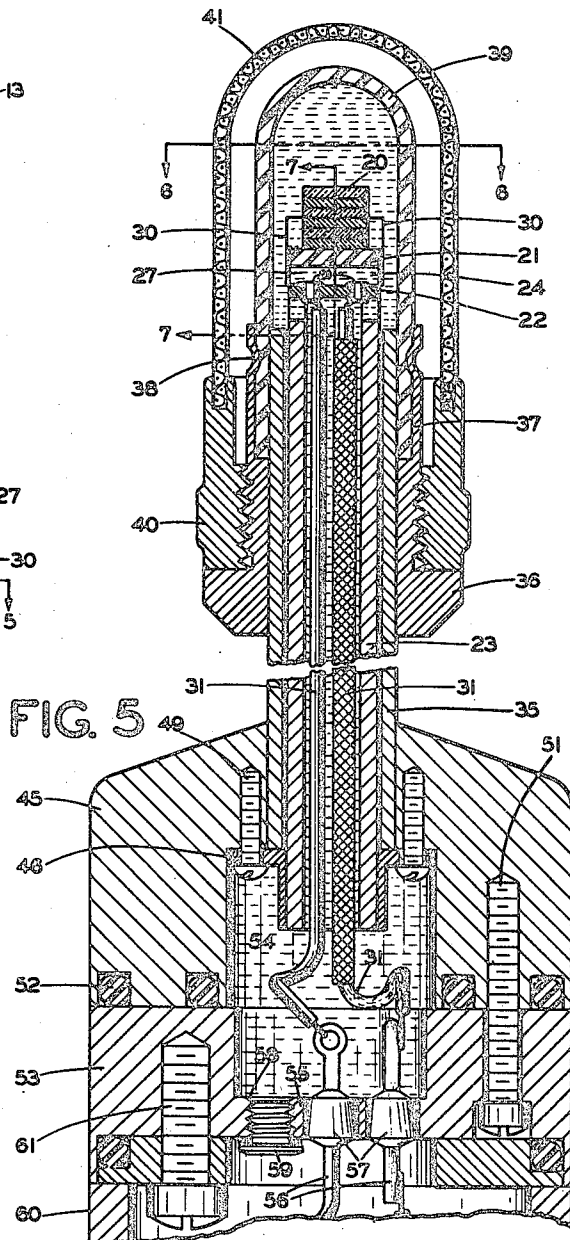


FIG. 5

INVENTOR.
HANS JAFFE.
BY
Eber J. Hyde
ATTORNEY

UNITED STATES PATENT OFFICE

2,490,216

PIEZOELECTRIC CRYSTAL

Hans Jaffe, Cleveland Heights, Ohio, assignor to
The Brush Development Company, Cleveland,
Ohio, a corporation of Ohio

Application June 17, 1947, Serial No. 755,167

6 Claims. (Cl. 171—327)

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This invention pertains to piezoelectric crystal means and more particularly to such means exhibiting a hydrostatic and a longitudinal piezoelectric effect of large magnitude.

By the term "hydrostatic piezoelectric effect" is meant the coupling between a pressure applied uniformly to all sides of a piezoelectric crystal body and an electric field created by this pressure in a certain direction in the crystal which is variously called the electric axis, the polar direction, or polar axis. This effect is reversible as the hydrostatically sensitive crystal, upon being subjected to an electric field parallel to the polar axis, is subject to a change in volume which can be communicated to a surrounding fluid medium in the form of acoustic waves. Piezoelectric effects due to hydrostatic pressure are mentioned at page 194 in the book *Piezoelectricity* by Walter G. Cady, published by the McGraw-Hill Book Company in 1946.

The particular advantages of the hydrostatic effect in the construction of microphones and related acoustic instruments have been described, and tartaric acid and cane sugar crystals have been used in piezoelectric pressure gages. The crystal tourmaline has been used in pressure gages but its use is severely limited by the small supply of natural crystals of useful quality as well as by the comparatively small value of its piezoelectric modulus for hydrostatic pressure, which modulus is $2.4 \cdot 10^{-12}$ coulomb/newton. The hydrostatic piezoelectric modulus for tartaric acid is about twice that of tourmaline but is still rather low for wide-scale practical applications, and the modulus for cane sugar is even lower than that for tourmaline.

The term "longitudinal piezoelectric effect" as used in this specification is intended to mean a piezoelectric coupling between an electric field component and a compressional motion parallel to the field component. Such longitudinal effects are well known for the crystals quartz and tourmaline, but these crystals have been obtained in commercial quantities only from natural sources which are limited in supply of high-grade material. Furthermore, the coupling for the longitudinal piezoelectric effect in quartz and tourmaline crystals is only about 0.10, which is a comparatively small value. Methods have been disclosed by which longitudinal piezoelectric crystal plates may be obtained from synthetic crystals such as Rochelle salt. Thus in United States Letters Patent No. 2,170,318, granted on August 22, 1939, to Walter G. Cady and in application for United States Letters Patent Serial

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Number 539,312, filed June 8, 1944, in the name of Hans Jaffe (now Patent No. 2,463,109, granted March 1, 1949), and assigned to the same assignee as the present invention, there are illustrated methods for obtaining crystals of primary ammonium phosphate and other crystals isomorphous therewith. However, with crystals belonging to the crystal classes of Rochelle salt and primary ammonium phosphate, these longitudinal piezoelectric plates are obtained only by cutting at an angle to the crystal axes. An analysis of the motion of such plates shows that only part of the applied electrical energy is employed to excite the crystal in its piezoelectric modes, and again only part of this mechanical excitation appears as a vibrational energy in the thickness mode of said plate. Aside from a reduction in the effective piezoelectric coupling, this condition leads to excitation of parasitic modes leading to complex vibration patterns.

It is an object of the invention to provide hydrostatic piezoelectric crystal means which does not exhibit any one of the several disadvantages of the previously known hydrostatic piezoelectric crystal means.

Another object of the invention is to provide piezoelectric crystal means having a high hydrostatic effect.

A further object of the invention is to provide hydrostatic piezoelectric crystal means for transducer use which will improve the signal-to-noise ratio of the transducer.

Still another object of the invention is to provide synthetic piezoelectric crystal means having high hydrostatic and longitudinal piezoelectric effects.

Yet another object of the invention is to provide piezoelectric crystal means for use in a wide-band tuned transducer.

In accordance with the invention, there is provided as an article of manufacture, a piezoelectric body composed, aside from any impurities, of a substance selected from the group consisting of lithium sulfate monohydrate, lithium selenate monohydrate and mixtures thereof, and having a pair of substantially electroducible surfaces substantially perpendicular to the Y axis of the crystal material.

Other objects and a fuller understanding of the invention may be had by referring to the following description, claims, and drawings.

Fig. 1 of the drawings is an isometric view of a typical crystal of lithium sulfate monohydrate ($\text{Li}_2\text{SO}_4 \cdot \text{H}_2\text{O}$); Fig. 2 is an isometric view of a plate cut from the crystal shown in Fig. 1 and

showing elements which may be obtained therefrom; Fig. 3 is a plan view of the plate cut from the crystal shown in Fig. 1 showing other elements which may be obtained therefrom; Fig. 4 is an isometric view of a stack of crystal plates; Fig. 5 is a sectional view, taken along line 5—5 of Fig. 6, through a transducer utilizing the crystal block shown in Fig. 4; Fig. 6 is a sectional view taken along line 6—6 of Fig. 5; Fig. 7 is a sectional view taken along line 7—7 of Fig. 5; and Fig. 8 is an isometric view of a detail of the transducer shown in Fig. 5.

The present invention pertains to crystal bodies cut from crystals of lithium sulfate monohydrate, $\text{Li}_2\text{SO}_4 \cdot \text{H}_2\text{O}$, lithium selenate monohydrate, $\text{Li}_2\text{Se} \cdot \text{H}_2\text{O}$, and mixtures thereof with particular orientations such that the bodies have a high longitudinal piezoelectric effect and exhibit a hydrostatic piezoelectric effect much larger than the effect for any heretofore known crystal body. The lithium sulfate monohydrate crystal is much superior to the selenate crystal due to the comparative costs of the raw materials from which the crystals are grown, and hence this invention is described as particularly adapted to the sulfate crystal, but it is to be understood that the invention includes the selenate crystal and also includes crystals grown from mixtures of lithium sulfate monohydrate and lithium selenate monohydrate.

If a hydrostatic piezoelectric crystal body is subjected to uniform hydrostatic pressure on all of its faces, no shearing stresses will be introduced into the crystal body. Accordingly, the piezoelectric moduli effective to determine the hydrostatic modulus are d_{21} , d_{22} , and d_{23} , if the polar axis is chosen as the Y coordinate axis in accordance with the nomenclature recommended by the Committee on Piezoelectricity of the Institute of Radio Engineers for the monoclinic polar crystal class, to which the lithium sulfate and selenate crystals here described belong. Crystallographic axis a , b , and c are chosen on the basis of the unit cell dimensions as determined by Ziegler (*Zeitschrift f. Kristallographie*, volume 89, 1932). The coordinate axes X, Y, and Z are correlated thereto in accordance with the nomenclature mentioned above. If the piezoelectric moduli d_{21} , d_{22} , and d_{23} do not cancel each other there is a resultant polarization in the direction of the polar axis which is termed the hydrostatic piezoelectric modulus.

The piezoelectric modulus d_{22} describes the strain (relative elongation) parallel to the Y axis produced by a unit electric field applied in the same direction. The moduli d_{21} and d_{23} indicate the strain parallel to the X and Z axes, respectively, produced by the unit electric field parallel to the Y axis. There is, further, a modulus d_{25} indicating the shearing strain around the Y axis produced by the unit electric field parallel to the Y axis. The same piezoelectric moduli are applicable to the direct piezoelectric effect where they describe the charge per unit surface area produced by a unit stress.

The piezoelectric moduli relating to an electric field in the Y direction of lithium sulfate are, respectively: $d_{21}=+1.5$; $d_{22}=+16.0$; $d_{23}=-4.0$ and $d_{25}=-5.2$ where the unit is 10^{-12} meters/volt or 10^{-12} coulomb/newton. The modulus d_{22} , describing the interaction between an electric field and a compressional motion both of which are parallel to the Y direction, is about seven times as high as the corresponding coefficient for the widely used quartz X-cut. The resonant fre-

quency of plates and bars vibrating in longitudinal motion in the Y direction is 2735 kc./mm. for the plate with air gap, and 2360 kc./mm. for the bar electroded on its end faces. From the latter value and the previously known density of the crystalline material which is 2.06, the elastic compliance for the Y direction is found as $21.7 \cdot 10^{-12}$ meter²/Newton. The dielectric constant of a Y-cut body of lithium sulfate monohydrate measured at frequencies below the resonance of such a body is 10.3. From these data, a piezoelectric coupling coefficient for the longitudinal vibration parallel to the Y axis excited by a field in the same direction is found to be 0.36. This is a remarkably high value for a piezoelectric coupling, being exceeded only by Rochelle salt where, however, the high coupling refers to a mechanical stress at right angles to the direction of the electric field. The advantage of high piezoelectric coupling between a mechanical stress and an electric field parallel to the stress for many applications such as the excitation or detection of ultrasonic waves, particularly in a liquid medium, in the frequency range of 100 kc. and above is well known. The extent of improvement possible by the use of Y-cut bodies of lithium sulfate monohydrate over quartz and other crystal plates whose coupling coefficient is low is better appreciated when it is realized that most piezoelectric underwater acoustic systems utilize crystal transmitters as well as crystal receivers. The coupling coefficient enters into the efficiency of each of these two transducers which means that the electrical output of the pickup transducer is dependent on the square of the coupling coefficient. Thus the output from such a system utilizing lithium sulfate monohydrate is about ten times as high as the output from a similar system utilizing quartz. In crystals of lithium sulfate monohydrate, piezoelectric effects are also observed for electric fields acting in the plane perpendicular to the polar axis but these additional piezoelectric effects are not of the hydrostatic or longitudinal type.

The crystal is stable, if protected from excessive humidity, at any temperature up to 75° C., but the crystal should be protected against loss of its water of crystallization if it must be exposed to higher temperature. In an atmosphere of suitable humidity, it is stable to 230° C. The hardness of this crystal is comparable to calcite and noticeably higher than that of Rochelle salt, primary ammonium phosphate and primary potassium phosphate.

The electric resistivity of the lithium sulfate crystal is extremely high, values up to 10^{12} ohm-cm. having been observed.

The dielectric strength of lithium sulfate Y-cuts is highly satisfactory. It was found, for instance, that a plate of 0.62 mm. thick withstood a D. C. voltage of 8000 volts.

The temperature coefficient of resonance frequency for these Y-cuts was found to be in the order of $-0.01\%/^{\circ}\text{C}$. The dielectric constant increases about 0.01% for one degree centigrade. The temperature dependence of the piezoelectric modulus d_{22} was too small to be measured but does not exceed $0.2\%/^{\circ}\text{C}$. The small value of all these temperature coefficients assures the operation of a lithium sulfate Y-cut transducer to be substantially unaffected by temperature variation in a normal operating range.

As has previously been stated, the piezoelectric modulus of tourmaline for hydrostatic pressure is

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$2.4 \cdot 10^{-12}$ coulomb/newton. The corresponding modulus for lithium sulfate monohydrate is obtained as $13.5 \cdot 10^{-12}$ coulomb/newton, this being the sum of d_{21} , d_{22} and d_{23} , which is nearly six times as high as for tourmaline. This means that a hydrostatic transducer device utilizing the new crystal body will not have to have as much electrical amplification as the prior art devices and will have a better signal-to-noise ratio.

In Fig. 1 there is shown a typical crystal of lithium sulfate monohydrate. The crystal is of the monoclinic crystal system and the polar crystal class, and may be either a "right" or "left" hand crystal. Illustrated is a "right hand" crystal, and the axes have been so defined that the sign of the most important piezoelectric modulus d_{22} is positive.

The invention and the relationships herein described pertain equally to "left" and "right" hand crystals with the provision that a right handed coordinate system is used for the right hand crystal, and a left hand coordinate system is used for the left hand crystal.

Hydrostatic crystal bodies and longitudinal expander bodies are obtained from the crystal by cutting blanks having faces perpendicular to the Y axis of the crystalline material, as shown by the crystal blank 10 in Fig. 1. For the fabrication of hydrostatic transducers, it is preferable that this crystal blank 10 be plate-like as the transducers made therefrom usually use a number of such plates cemented together to increase the capacity, as is known to the art. While the greatest use by far is for plate-like bodies it is sometimes desirable to cut cubical and rod-like bodies.

A crystal plate to be cut from the blank 10 may have any configuration, and if it has straight sides they may be at any desired angle to the X and Z axes of the crystalline material. Where a large plate is wanted and the mother crystal is limited in size, a long edge of the crystal plate should lie in the (100) face of the crystal which is usually the largest face of the grown crystal. Such a plate is identified by the reference character 11 in Fig. 2. If a very thin plate or a narrow crystal bar is wanted, it is advantageous to cut the crystal so that one pair of its edge faces are parallel to the plane of the (101) face of the crystal as this plane is a cleavage plane and the danger of undesired fracture of the crystal plate is reduced if there is not a pair of side faces making a small angle with the cleavage plane. Reference character 12 in Fig. 2 identifies the (101) face which is parallel to the cleavage plane. In many cases a crystal plate with a rectangular outline may not be necessary, in which event it may be advantageous for the sake of manufacturing ease and economy to make one pair of side faces parallel to the (101) face and another pair parallel to the (001) faces. These side faces will then be at an angle of about 69 degrees to each other, as shown by plate 9 in Fig. 3.

Other orientations of the side edges of Y-cut piezoelectric plates of the lithium sulfate crystal may be chosen to achieve particular operating characteristics. Where it is desired to obtain a transducer body, the lowest piezoelectrically excited resonance frequency of which is as high as possible for a given dimension of the body, the side edges should be so oriented that the face shear mode of vibration is not excited. Choosing a coordinate axis X' parallel to one side edge of the desired plate, the condition for vanishing face

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shear excitation is $d_{25}' = 0$. From the measured piezoelectric coefficients given earlier, it is found that this condition is fulfilled if the X' axis includes an angle of plus (+) 68° with the X axis. The plus (+) sign indicates that the X' axis lies in the quadrant between the -Z and +X directions.

In some applications it is desirable that the pattern of motion set up by the crystal plate in a surrounding medium be as uniform as possible around the Y axis of the crystal. In this case it is desirable to orient the side edges of the crystal plate so that the expansion along the side edges of the crystal is equal for an excitation by a field in the Y direction. If an axis X'' is placed parallel to a side edge of such a desired plate, the condition for obtaining this equality of expansion is $d''_{21} = d''_{23}$. With the measured data of the piezoelectric moduli for lithium sulfate, it is found that the X'' axis must make an angle of about $+23^\circ$ with the X axis.

The two rectangular plates described in the preceding paragraphs have their edges related to each other by a rotation of 45° around the Y axis and are in a relationship akin to that of expander plates and shear plates of Rochelle salt crystals.

Other orientations of the side edges of the Y-cut plate may be derived from the requirement of minimum coupling between lateral and shear expander modes. Further orientations may be derived from the requirement that plates cut from one type of lithium sulfate crystal, either "right" or "left" and joined on their Y faces in opposite polarity such as required for construction of transducer 20, shall have identical position of their thermal expansion axes to reduce cracking of the plates due to thermal changes.

The bodies cut from the crystal shown in Fig. 1 are electroded by applying to those faces which are substantially perpendicular to the Y axis thin layers of graphite, gold, silver, or any other electrically conducting material indicated by reference character 13, as is well known to the art. Leads 14, 15 are connected to the electrodes to translate the electric charges produced by a field parallel to the polar axis or by the application of an external stress to the crystal body with a component parallel to the Y-axis.

Figs. 4 to 7 illustrate a practical transducer design, the details of which are described and claimed in application Serial No. 755,128, filed concurrently herewith in the name of Harry B. Shaper and assigned to the same assignee as the present application, utilizing a plurality of Y-cut plates 11 of lithium sulfate monohydrate connected together in face-to-face relationship with opposite polarity to form substantially a cube 20 of crystalline material. Leads 14 and 15 respectively, are connected together so that the several crystal plates 11 are in parallel. This construction reduces the voltage output and increases the capacity of such a transducer used as a microphone compared to a solid cube of crystalline material, as is known to the art. It is not essential for some transducers that the plurality of plates 11 form a cube but it is advantageous where a non-directional transducer is desired for use in a frequency range where the length of the wave in the surrounding medium is comparable to the greatest dimension of the plurality of plates. These advantages are also known to the art.

The complete assembly of Fig. 5 is designed

particularly for use in water from low audio frequencies up to about 100,000 cycles per second. The crystal cube 20 is connected by an adhesive such as rubber cement to the top surface of a block 21 of rubber or the like having an acoustic resistance comparable to the acoustic resistance of water. The bottom surface of the rubber block 21 is cemented to the top surface of a cap 22, and the bottom surface of the cap 22 is cemented to a long hollow tube 23. The cap 22 and the tube 23 may be formed of any hard material, such as a phenolic condensation product such as that sold under the trade name "bakelite," and any suitable cement may be used to secure them together. The cap 22, shown isometrically in Fig. 8, comprises a base portion with integral upstanding edge portions 24 to the top surface of which the rubber block 21 is cemented. Thus it has a broad and deep groove 27 extending across its top surface, leaving a thin section 28 of material at the bottom of the groove. Rivets 29 extend through this thin section 28 for holding lead connection lugs 30 and to which wires 31 from an amplifier are connected. The lugs 30 extend out of the groove 27 and are soldered to the crystal leads 14, 15 to complete a circuit from the crystal 20 to the amplifier (not shown). The broad groove 27 also permits sound waves to impinge directly onto the back face of the block 21 of rubber through which they pass to the crystal block 20 with minimum energy loss. The tube 23 which supports the crystal 20 is positioned inside a hollow metal tube 35. Near the upper end of the metal tube 35 there is an externally threaded mounting member 36 which is soldered to the metal tube, and the mounting member 36 includes at its upper end a thin ring portion 37 which surrounds the upper end of the metal tube 35 with a small space between them. The lower edge of a rubber cover member 39 is positioned in the space between the ring 37 and the upper end of the metal tube 35, and the ring 37 is grooved at 38 to cause the rubber cover to be tightly connected to the tube 35. An internally threaded collar 40, to which is connected a screen 41, is screwed into engagement with the mounting member 36 to position the screen 41 around the rubber cap 39. The screen is sufficiently stiff to protect the rubber cover 39 and the crystal within from damage. A base member 45 is soldered to the tube 35 near the lower end of the tube to effect a fluid-tight seal. The base member 45 is in the form of an inverted cup having a broad flat lip. Connected to the base of the tube 23 by a screw (not shown) is a collar, at the upper end of which is a broad circular ring 46. Screws 49 extend through the ring into the base 45, thereby positioning and holding the tube 23 within the metal tube 35. A cup-shaped cap member 53 having a central partition 55 is connected against the broad lip of the base 45 by means of a plurality of screws 51 (only one of which is shown), and fluid sealing means 52 are positioned therebetween in order to prevent liquid from leaking out between the base 45 and the cap 53. Within the hollow 54, formed by the cup in

the base 45 and the cup in the cap 53, the crystal lead wires 31 are connected to two wires 56 which extend through the partition 55. Glass seals 57 insulate the wires 56 from the cap 53 and provide a fluid-tight seal around the wires. The partition 55 has a threaded opening 58 through it, and after the transducer has been assembled a fluid, such as castor oil, is put in through the opening to completely fill the volume inside the rubber cap 39, inside the hollow tube 23, between the tube 23 and the tube 35 and in the enclosure 54. A plug 59 is then screwed into the opening 58 to close it. The aforescribed assembly constitutes the transducer proper, and to the transducer proper there is connected by screws 61 a preamplifier housing 60 the details of which are described in the aforementioned Shaper application.

While the invention has been described with a certain degree of particularity it is to be understood that numerous changes can be made in the parts and their arrangement without departing from the spirit and scope of the invention as hereafter claimed.

I claim as my invention:

1. As an article of manufacture, a piezoelectric crystal body composed, aside from any impurities, of a substance selected from the group consisting of lithium sulfate monohydrate, lithium selenate monohydrate and mixtures thereof, and having a pair of substantially parallel electroducible surfaces substantially perpendicular to the Y axis of the crystal material.

2. An article of manufacture as claimed in claim 1 in which the crystal body is in the form of a plate having electroducible major faces substantially perpendicular to the Y axis of the crystal material.

3. As an article of manufacture, a crystal plate as claimed in claim 2 which is quadrilateral in outline with one pair of side surfaces parallel to the Z axis of the crystal material.

4. As an article of manufacture, a crystal plate as claimed in claim 2 which is quadrilateral in outline with one pair of side surfaces parallel to the cleavage plane of the crystal material.

5. As an article of manufacture, a crystal plate as claimed in claim 2 which is quadrilateral in outline with one pair of side surfaces at an angle of substantially +68 degrees to the X axis of the crystal material.

6. As an article of manufacture, a crystal plate as claimed in claim 2 which is quadrilateral in outline with one pair of side surfaces at an angle of substantially +23 degrees to the X axis of the crystal material.

HANS JAFFE.

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The following references are of record in the file of this patent:

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